

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Application No: 10/537,651
Applicant: Lindall et al.
Filed: October 31, 2005
Title: CATALYST AND PROCESS
T.C./A.U.: 1793
Examiner: McDonough, James E.
Confirmation No.: 5162
Docket No.: JMYS-128US

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

DECLARATION OF CALUM HARRY MCINTOSH PURSUANT TO 37 CFR §1.132

I, Calum Harry McIntosh, being duly warned that willful false statements and the like are punishable by fine or imprisonment or both, under 18 U.S.C. §1001, and may jeopardize the validity of the patent application or any patent issuing thereon, state and declare as follows:

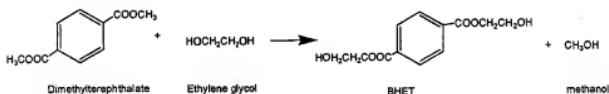
1. All statements herein made of my knowledge are true and statements made on information and belief are believed to be true.
2. I received my Bachelor of Science degree in Chemistry at Heriot-Watt University, located in Edinburgh, in 1989. I received my Master of Philosophy degree in Organometallics from the same university, in 1991.
3. I have been employed by Johnson Matthey PLC and in the businesses of Imperial Chemical Industries PLC and Tioxide Specialties Ltd, which are currently owned by Johnson Matthey PLC, for 18 years. I am currently a Technical Sales Manager for the Refineries business of Johnson Matthey, a position I began on January 1st 2009. Prior to this current appointment, from 1999 – December 31st 2008, I was Technical Development Manager in the VERTEC polymers business of Johnson Matthey, responsible for the

development, commercialization and manufacturing processes of a number of organometallic products, in particular organotitanates for use as catalysts for polyester manufacture. A copy of my *curriculum vitae* is attached hereto as Exhibit A.

4. I was working as a Technical Manager in the polyester catalysis research group in which the invention to be protected by this patent application was made during and after the date of the invention. This is the same research group in which the inventions described in European Patent No. 0 812 818 (Ridland et al.) were made.
5. I have read, and am familiar with, the contents of this patent application, the Office Action dated January 29, 2009, and the cited reference, European Patent No. 0 812 818 (Ridland et al.). I understand the nature of the obviousness rejections at issue in this application. In particular, the Office has asserted one of ordinary skill in the art at the time of the invention would have determined the optimal ratio of base to acid through routine experimentation in the art in an effort to optimize the catalyst performance. This declaration addresses these issues.
6. By way of background, polyester polymer such as polyethylene terephthalate (PET) and polybutylene terephthalate (PBT) is typically manufactured using two main process stages. In the first stage, a dicarboxylic acid such as terephthalic acid, or an ester of the acid, usually a dimethyl ester, e.g. dimethyl terephthalate is reacted with a dihydric alcohol. When the required polyester is PBT then the dihydric alcohol used is 1,4-butanediol (BDO). When the required polyester is PET then the dihydric alcohol used is ethylene glycol. Other compounds may be used in addition to the principal alcohol and acid components to form various copolymers and modify the polyester properties. The reaction between terephthalic acid and the glycol, displacing water from terephthalic acid. The following reaction scheme shows the direct esterification of terephthalic acid with ethylene glycol to form bis(hydroxyethyl)terephthalate (BHET).



When an ester of terephthalic acid, e.g. dimethylterephthalate, is used as the starting material instead of terephthalic acid itself, then the ethylene glycol displaces the alcohol (e.g. methanol) from the starting ester.



The water or methanol is removed from the reaction mixture. Processes starting from terephthalic acid and from dimethylterephthalate are both widely used in the commercial production of polyesters. The reaction, normally takes place at a temperature higher than 200 degrees centigrade and continues until the evolution of water or methanol has ceased.

In the second stage of the polyester manufacture, the BHET is polycondensed by heating the reaction mixture under reduced pressure to a temperature higher than the melting point of the polyester. When ethylene glycol is used as the dihydric alcohol, it is released from the BHET and removed from the reaction mixture as it is formed.

Stage 2: Polycondensation (PC)



A catalyst may be added to a polyester manufacturing process at any of several stages in the process. When the catalyst is added at the start of stage 1 of the process, it is often added as a mixture with the ethylene glycol or other dihydric alcohol. A liquid catalyst can be thoroughly mixed with the glycol. When added at a later stage, e.g. to a liquid reaction mixture such as

molten BHET, then the use of a liquid catalyst enables rapid and thorough distribution throughout the reaction mixture and avoids undue deactivation of the catalyst before it is used in the reaction. For these reasons, it is highly desirable to use a liquid catalyst in polyester production.

7. In 2001, The Research and Development team of the VERTEC business of Johnson Matthey, of which I was a part, wished to make a catalyst suitable for use in making polybutylene terephthalate (PBT). As previously explained, PBT is a polyester made from dimethyl terephthalate and 1,4-butanediol (BDO). BDO is used in place of ethylene glycol in the above process description and reaction schemes but otherwise the reaction is broadly similar. The desire was to make a liquid catalyst which was miscible in BDO and which contained no other polyol which might be incorporated into the finished polyester because this may affect the properties of the polyester product. The decision was made to use the known recipes of the Ridland patent, but to substitute the ethylene glycol used in the Ridland catalysts with BDO.
8. A composition was prepared in the laboratory at Billingham by reacting citric acid with titanium isopropoxide, removing the isopropanol formed in the reaction and then partially neutralizing the acidic groups of the citrate with aqueous sodium hydroxide. The product was then mixed with BDO and heated to remove isopropanol and water. The detail of the experiment is described in Example 1 of this patent application. The molar ratio of base : acid was 1.2 : 1 because this was the amount known to be effective from the Ridland patent and used as a standard recipe by our research team. It was found that the resulting composition was not a mobile liquid, as required, but instead formed a gel. For the reasons given above, the resulting product is not suitable for use as a catalyst for polyester manufacture. The polyester research team, then led by Charles Lindall, named as Inventor on this patent application, subsequently tried to make the catalyst containing BDO using less base than had previously been used in our standard catalyst recipes and found that a hydrolytically stable, liquid catalyst composition could be made in this way.

9. I note that the quotation from the Ridland patent in the Office Action is not complete, in that the subsequent sentence reads "In the case of citric acid (a tribasic acid), the preferred amount [of base] is in the range 1 to 3 moles base per mole of 2-hydroxy acid". The preparation of the new catalyst with BDO used citric acid. The improvement of the recipes disclosed in the Ridland patent, using less base than the amounts disclosed explicitly in the Ridland patent, led to the result that a liquid product could be manufactured using BDO as the dihydric alcohol. This is commercially significant in that it enabled a catalyst to be manufactured which is useful in the manufacture of PBT.
10. The new recipe as described in this patent application was then tried in a catalyst composition containing ethylene glycol. Our standard catalyst recipes, based on the inventions disclosed in the Ridland patent, had used ethylene glycol but had contained sodium hydroxide base at an amount calculated to provide a molar ratio of base : acid of about 1.2:1. Once again, a stable liquid catalyst composition containing less than 0.8 moles of base per mole of citric acid was formed. This composition is described in this patent application as Example 14 and will be referred to hereinafter as "New PET catalyst".
11. Some customers using our commercial catalyst had noticed that, when forming fibers from polyester made with our standard catalyst, the fibers were susceptible to breaking during processing. Analysis of the fiber samples in our laboratories showed the presence of localized concentrations of sodium and titanium associated with the polyester at the break points. We had not been aware of the potential role of sodium in causing fiber breakage and had not expected to find this result. In an attempt to solve this problem, the research team carried out an experiment making PET using the New PET Catalyst. When used to form polyester fiber it was found that the fibers broke less often during processing than had been the case using the standard catalyst containing higher levels of base.
12. As part of the research team activities, I carried out thermal analysis on the PET polyester made using the New PET Catalyst. The thermal analysis experiment is described in this patent in Example 20. I was surprised to find that the sample of polyester made using the New PET Catalyst showed an

onset of crystallization (T_{n_0}) 5 degrees centigrade higher than the T_{n_0} of polyester made using the standard catalyst. These results are shown in Table 4 of this patent application and indicate that the polyester made using the New PET Catalyst crystallizes more slowly. The significant rise in crystallization temperature was surprising and would not have been expected to result from reducing the amount of base used in the catalyst from a molar ratio of 1.2 (NaOH) : 1 (citric acid) to 0.67(NaOH) : 1 (citric acid).

13. The raised crystallization temperature in the PET made using the New PET Catalyst is commercially important. This is because polyester resin is made into commercial articles such as bottles, films and fibers by forming those articles from molten polyester. When the polyester has solidified from the melt, it is further processed by orienting the polymer molecules to form dimensionally stable and strong articles. This processing, which includes stretching bottles and films and drawing fibers, must take place at a temperature above the glass transition temperature (T_g) of the PET but below the crystallization temperature of the PET because when the polymer is crystallized the oriented structure in the polymer is set. With the slower crystallization exhibited by polyester made using the New PET Catalyst, the temperature "window" within which the PET processor can form and orient the polyester articles is significantly widened so allowing more processing to take place before the polymer crystallizes. This is a significant benefit for a fabricator of polyester articles. This effect was wholly unexpected. I would not have anticipated being able to produce this effect, and thereby affect the thermal processing properties of polyester in such a commercially significant manner, from my knowledge of the background technology as described in the Ridland patent.



Calum Harry McIntosh

Date: 28/05/2009

Appln. No.: 10/537,651
Office Action of January 29, 2009

JMYS-128US

Attachments: Exhibit A - Curriculum Vitae

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EXHIBIT A

To the

DECLARATION OF CALUM HARRY MACINTOSH PURSUANT TO 37 CFR §1.132



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Training and Personal Development

2007 -	WinLims/SAP interface training
2005 - 2007 Johnson Matthey Plc	SAP training <i>Global Business Culture</i>
2004 Johnson Matthey Plc	Quality Auditing <i>Advanced presentation skills</i>
2002 Johnson Matthey Plc	Industrial Catalytic Processes, Diploma (University of Liverpool)
2001 ICI Syntex	Competition Law <i>Professional Selling</i>
1989 - 1991 University of Heriot Watt 1985 - 1989 University of Heriot Watt	Master of Philosophy (MPhil), Organometallics <i>3 publications</i> Bachelor of Science, Chemistry (BSc Honours) <i>2 publications</i>
Experience 2009 -	Johnson Matthey Plc, Billingham <i>Refineries Technical Sales Manager (Middle East Region)</i>
2007 - 2009	Johnson Matthey Plc, Billingham <i>Global Technical Development Manager for VERTEC growth/ Site Analytical Manager/Production Support</i> <ul style="list-style-type: none">◆ Delivered strategy for maximising manufactured product quality◆ Extending the capability of the analytical function in line with corporate strategy and sustainability◆ Maximising QA over QC through SPC/SQC programs◆ Delivered OEE quality figures surpassing business targets◆ Through continuous improvement programs developed and delivered a request for analysis/report procedure◆ Developing programs optimising synergies between manufacturing and technical functions
2003 - 2007	Johnson Matthey Plc, Billingham <i>Business Development Manager for VERTEC growth</i> <ul style="list-style-type: none">◆ Developed and implemented technology platform in line with commercial plan◆ Developed Intellectual Property (IP) strategy/management plan◆ Developed strategic alliances with leading technology providers to the global polyester industry

- ◆ Commercialised 3 new products on 6 world scale polyester lines increasing NRs by 50%
 - ◆ Obtained 3 new patents
 - ◆ Delivered presentations to Asian, European and American polyester congresses
 - ◆ Represent JM on EU polyester recycling association – PETCORE
- 2001 - 2003
- Johnson Matthey Plc, Billingham**
Technical Development Manager for VERTEC growth
- ◆ Developed 2 new products and obtained 3 new patents leading to 2 industry awards for innovation
 - ◆ Managed technology roll-out seminars in Asia leading to 3 new customers increasing NRs by 30%
 - ◆ Delivered laboratory analytical and technical service capability
 - ◆ Delivered technology training package and rolled-out to agents and distributors globally
 - ◆ Delivered improvements in product manufacture leading to 50% reduction in batch times and 30% reduction in costs to manufacture
 - ◆ Implemented long term development plans for all reports
- 1999 - 2001
- ICI Synetix, Billingham**
Technical Manager for core polymers business
- ◆ Delivered global marketing plan for the launch of 2 products into the plasticiser market through market research, value chain analysis and customer feedback leading to 10% increase in sales
 - ◆ Led a Customer Service technical team and developed the Technical Service capability
- 1994 - 1999
- Tioxide Specialties Ltd, Billingham**
Chemist : New Product Development
- ◆ Delivered extensive report on Industrial plasticizers market to establish product/technology opportunities and SWOT analysis on TSL offer
 - ◆ Converted 3 world scale plasticizer plants from incumbent catalyst technology to TSL technology
 - ◆ Compiled a Worldmarket survey on plasticizer producers and technologies employed
 - ◆ Delivered business literature outlining technology offer in areas of plasticizers and acrylates
- 1991 - 1994
- Johnson Matthey Plc, Hertfordshire**
Chemist : New Product Development
- ◆ Attended graduate business training program to allow commercial awareness and best business practices
 - ◆ Achieved award for best graduate project
 - ◆ Developed optimised formulations for the decorative coatings market
 - ◆ Developed a new manufacturing route for gold powder technology and therefore all products based on gold
- 1991
- Caltex Oil, Brisbane, Australia**
Analytical Chemist
- ◆ Working holiday – analysing lube oils and jet fuel for conformitydevelopment